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LOCKHEED MARTIN

G. Wilson Horde
Counsel

August 30, 1999

Mr. Edward B. Blackwood
Director, Radiological Protection
Bechtel Jacobs Company LLC
761 Veterans Avenue
Kevil, Kentucky 42053

Dear Ed:

Attached herewith please find the three reports that Brad Graves referred to when he was interviewed by you and Bill Cooper last Friday, August 27.

I am attaching a memo from Graves to A. H. Jefferies dated April 10, 1990; a memo from Graves to C. W. Walter dated April 23, 1990; and a memo dated March 11, 1960, which appears to be directed to Drs. C. L. Dunham and H. D. Bruner. The latter has some underlining on the memo, and in certain places it is difficult to read. The copy from which these were made was in the same condition. I am referring to Pages 3 and 4, and more particularly on the right-hand side of those two pages. If a better copy is located, I will be happy to forward that to you also if you desire.

Sincerely,

G. Wilson Horde

GWH:tlm

Attachments

c w/o att: M. B. Graves

MARTIN MARIETTA

Internal Correspondence

MARTIN MARIETTA ENERGY SYSTEMS, INC.

April 10, 1990

A. H. Jefferies

Paducah Gaseous Diffusion Plant (PGDP)
Evaluation of Transuranic Activity Levels

It has been understood by current Health Physics management that the transuranic materials at PGDP were of minimal quantity and located in very well defined waste storage areas. It was also understood that negligible amounts of transuranic contamination was located in general plant areas; however, the definable levels of activity posed no significant impact on the Health Physics program operations.

On March 22, 1990, while moving waste drums in building C-746-Q, a 55 gallon drum containing transuranic waste was spilled which resulted in a loss of approximately 20 gallons of liquid. In an effort to evaluate the spill from a radiological control point of view, the Health Physics Department requested contents of the drum. Calculations were performed to ascertain activity levels of the spill. Contents of the drum were reported as follows:

Americium 241	7.200×10^3 dpm/ml
Plutonium	5.700×10^4 dpm/ml
Neptunium 137	4.520×10^4 dpm/ml
Depleted Uranium (0.6%)	-3.600×10^4 dpm/ml
Technetium 99	$(1.625 \times 10^6$ dpm)/ml
Nitric Acid	M unknown PH=1

The spill area in C-746-Q was evaluated and radiological survey data was obtained. Several wipe surveys were taken and the Radiochemistry Laboratory was requested to perform a gross gamma scan on the wipe smears to ascertain the presence of transuranic activity. Radiological survey data is summarized on the attached chart.

As stated earlier, the presence of transuranic elements at PGDP were presumed understood. However, based upon the activity levels associated with the spill on results of the initial radiological survey information, the Health Physics Department began questioning the validity of prior information concerning transuranic activity at PGDP.

Immediate action was taken to investigate known areas containing transuranic activity and formulate a plan to reevaluate areas, processes, and radiological work which, in the past, rendered exposure of transuranic isotopes to personnel. Areas of PGDP to be monitored in support of transuranic isotope evaluation are as follows:

1. C-310 Cell Floor
2. C-400 Compressor Shop
3. C-400 Convertor Maintenance Area and Pulverizer
4. C-409 Convertor Maintenance Area
5. C-720 Compressor Shop
6. C-410 Ash Receiver Area
7. C-420 Ash Receiver Area
8. C-340
9. C-746-B Storage Area
10. C-335 Sample Manifolds Building

In addition to survey locations, a radiochemical transuranic evaluation will be required for all surveys of areas and equipment with transferable activity greater than 50,000 dpm/100 cm^2.

All smear surveys in plant locations specified will be analyzed for plutonium 239/240 and neptunium 237. A representative survey will be taken in each location. Any transferable survey, regardless of plant location, indicating levels of greater than 50,000 dpm/100 cm^2 will receive gross gamma scans by the Radiochemistry Laboratory to determine the presence of transuranic activity.

All radiological survey data will be transmitted to Health Physics Department management. Once representative data is obtained, decisions will be made as to further evaluations or Health Physics program changes.

M. B. Graves, C-743, PGDP, (6411)

MBG:psr

Attachment

cc/att: T. L. Barron
A. J. Potter
C. W. Walter
File - RC

RADIOLOGICAL SURVEY DATA

	Gross Alpha	Gross Beta	Uranium	Np	Uranium Enrichment
Floor Areas					
Not Affected by Spill	0 to 40 dpm	0 to 560 dpm	(not analyzed) (less than 100 dpm alpha)		
Affected by Spill	40 to 1,360 dpm	140 to 50,000 dpm	43 to 1,750 dpm	6 to 2,830 dpm	0.4 to 0.7%
Drum Surfaces					
In Diked Area	0 to 170 dpm	180 to 900,000 dpm	0 to 1,226 dpm	33 to 1,509 dpm	0.6%
UF_4					
From Broken Drum	970 dpm	7,000 dpm	2,700 dpm	16	0.2%

MARTIN MARIETTA

Internal Correspondence

MARTIN MARIETTA ENERGY SYSTEMS, INC.

April 23, 1990

C. W. Walter

Continued Transuranic Evaluation

Based upon the transuranic activity levels associated with the drum spill in building C-746-Q, reference memo entitled "Evaluation of Transuranic Levels" dated April 10, 1990, several actions have been taken to ensure timely evaluation of general transuranic levels at Paducah Gaseous Diffusion Plant (PGDP). To date actions are as follows.

Building C-746-Q spill site was isolated from all personnel until Health Physics evaluated the site.

Building C-746-Q spill site was decontaminated using continuous Health Physics job coverage. Decontamination attempts were only marginally successful.

Building C-764-B storage area was isolated from all personnel until Health Physics evaluated the site.

All personnel involved in the spill incident were evaluated for possible uptake using both in vivo and bioassay methods.

An investigation was conducted to include the perusal of historical radiological survey data and past plant operations to determine past transuranic activity levels and likely locations suspect to transuranic activity.

All technicians were requested to perform transferable surveys in key locations and send samples to the laboratory for analysis.

At approximately 3 p.m. on April 18, 1990, additional radiological data was obtained from the Radiochemistry Laboratory. Subject radiological data indicated positive transuranic activity at the spill site, storage location, and process systems. Survey data is summarized as follows.

Location	Transuranic Activity (dpm/100 cm2)*
C-746-B Storage Area	0 to 4,000
C-746-Q Storage Area (Post Decontamination)	0 to 2,820
C-337 Unit 1, Cell 2, Stage 2	0 to 1,284

* Laboratory data is from gross gamma scan of Neptunium (Np^{237}). It should be noted that additional transuranic activity is expected from Plutonium 239/Plutonium 240 ($Pu^{239/240}$) and Americium (Am^{241}). These isotopes cannot be evaluated using gamma scanning techniques.

A meeting was held at 8:30 a.m. on April 19, 1990, to discuss afore mentioned findings. Those in attendance were:

C. W. Walter	Health and Safety Division, Manager
C. R. Beverly	Quality and Technical Services
P. D. Wooldridge	Quality and Technical Services
T. L. Barron	Contamination Control Program Office
M. B. Graves	Health Physics Department
A. H. Jefferies	Health Physics Department

Through general discussion, concerns were raised as to the validity of the survey information. It was stated the amount of activity in the samples was not of sufficient level to ensure accuracy due to instrumentation lower limit of detection capabilities. It was also stated the limit of error could be as much as 100 percent. The decision was made to take additional samples of larger surface areas to increase the level of certainty of the survey information. Based upon conversation, the Health Physics Department is concerned about the ability of the Radiochemistry Laboratory to support our general requests, particularly if the presence of transuranic activity is substantiated.

A staff meeting was held at 9:30 a.m. on April 20, 1990, with the Health Physics technicians. The technicians were requested to obtain additional samples using masslinn cloth over large surface areas. It was stated radiochemical analysis of additional samples will be available April 26, 1990. As stated by the Health Physics Department in the meeting April 19, 1990, the validity of the initial data is indeed in question and should be substantiated. Based upon recently reviewed historical data and increased knowledge of plant operations, the Health Physics Department must take immediate action to control the likely presence of transuranic activity in the workplace regardless of questionable data.

Immediate action should include but not be limited to the following.

Evaluation of Radiochemistry Laboratory Capabilities

Radiological survey data must be made available in a timely fashion. It is understood the laboratory capability to support this effort is very limited due to manpower, analysis requirements, and the number of samples to be analyzed. Due to the presence of transuranic contamination, analysis requirements will increase dramatically. Initially, all air samples must be analyzed for transuranic activity. The entire PGDP site must be evaluated for transuranic surface contamination levels which requires specific laboratory support. In order to eliminate respiratory protection requirements, airborne radioactivity concentrations must be determined. The Health Physics Department will be able to make initial evaluations by obtaining gross alpha counts; however, the derived air concentration guides for uranium versus

Sent to Nyrath by Courier 5/23

transuranic activity are largely different and will require special analysis. It is strongly suggested PGDP attain outside radiochemical support capabilities to include the acquisitions of additional radiochemical instrumentation. It is understood that the amount of support needed will be difficult to obtain. Therefore, the Health Physics Department's protective requirements for radiological work will be extremely conservative.

Air Sampling/Monitoring Requirements

The primary personnel exposure pathway will be inhalation of radioactive material. The Health Physics Department's current capabilities for surveying airborne radioactivity concentrations is a documented weakness. The presence of transuranic activity at PGDP only compounds the problem. Immediate action will be to eliminate any unnecessary surface abrasion of radioactive contaminated material. The Health Physics Department will exercise work stoppage authority on all work involving burning, welding, grinding, or the breach of primary systems until continuous Health Physics job coverage can be provided. Air monitoring/sampling capabilities will be increased by acquiring additional samplers/monitors and annular kinetic impactors. A request was made to Portsmouth to make impactors available until PGDP can acquire their own. The Health Physics Department will relax protective requirements only when documentation and radiological data supports the action.

Unrestricted Release Criteria

Upon immediate verification of the presence of transuranic activity, the release criteria for the location, area, building, etc., will be commensurate with those limits set forth in DOE Order 5480.11 for transuranics. Until further notice, all shipments off-site will be surveyed assuming the presence of transuranic activity and the limits associated with same shall apply.

Instrumentation

Upon immediate verification of the presence of transuranic activity, instrumentation capable of detecting 300 dpm/100 cm^2 will be provided to allow personnel monitoring from the area. Survey instrumentation capable of detecting 300 dpm/100 cm^2 fixed contamination will be used to allow for release of all other items. All transferable contamination samples will be counted with the Health Physics laboratory equipment and a limit of 20 dpm/ 100 cm^2 will be used.

Contract Health Physics Support

The presence of transuranic activity severely impacts the Health Physics Department's capabilities to apply all aspects of radiological control at PGDP. It is imperative for future operations at PGDP to quickly quantify and qualify the transuranic problem. It is suggested that PGDP acquire the services of contractual Health Physics personnel in support of this effort.

A professional consultant with significant experience in dealing with transuranic isotopes should be obtained to assist in the evaluation process and upgrade of the radiological controls program. An adequate number of contract Health Physics technicians should be used to gather survey information and provide supporting documentation of the overall effort.

This document provides the minimal technical basis and/or actions required for the evaluation of transuranic isotopes at PGDP. This document should not be considered all encompassing. Upon your perusal of same, it is requested that comments on this program be made available as soon as possible. A more detailed supporting document on the Health Physics Department action plan is forthcoming.

If you have any questions or suggestions, please contact me.

M. B. Graves, C-743, PGDP (6411)

MBG:psr

cc: T. L. Barron
A. H. Jefferies
File - NoRC

March 11, 1960

Files

THRU : C. L. Dunham, M.D., Director, Division of Biology and Medicine
H. D. Bruner, M.D., Chief, Medical Research Branch, Division of Biology and Medicine

NEPTUNIUM^{237} CONTAMINATION PROBLEM, PADUCAH, KENTUCKY, FEBRUARY 4, 1960

SYMBOL : :MDB

Those contacted were Mr. B. Stiller and Mr. Nietsche of the AEC, Paducah Area Office and Dr. Neal Ward and Messrs. Don Levin, Ed Cain and R. G. Brown of Carbide. Mr. Joe Lenhart of ORO0 came up from Oak Ridge and took part in the discussions.

Np^{237} seems to be found only in reclaimed feed material provided by Hanford and therefore it is not a problem for the other separations plants.

It is produced by one or both of the following reactions:

(1) $_{92}U^{238}$ (N, p.2n) $_{93}Np^{237}$

(2) $_{92}U^{235}(n,\gamma)\ _{92}U^{236}$; $_{92}U^{236}(n,\gamma)\ _{92}U^{237}$; $_{92}U^{237} \xrightarrow[7\ \text{days}]{B^- + \gamma}\ _{93}Np^{237}$

This reclaimed U from Hanford now has about 0.05 g of Np/ton of U. The presence of Np was recognized as far back as 1957. At one time during 1958 this feed material had as much as 1 gram/ton but it has been lower lately because Hanford is extracting the Np for other purposes; it would not pay Paducah to try to remove completely this residue and in any case their problem comes from the Np already in the cascade units which now must be taken out, repaired, restored and put back in the systems.

The uranium comes to Paducah as UO_3 which is then reduced to UO_2 and treated with HF to get the green salt UF_4; this is then refluxed with F_2 gas in a hot cyclone type of pipe. The volatile UF_6 so formed goes out the top to be cooled and stored in the solid state in metal "bottles." All contaminants supposedly drop to the bottom of this cyclone pipe and are removed as "ashes," but it appears that Np has sufficiently similar chemical and physical properties to follow along with the UF_6 as NpF_6. The "ashes" show about 15% of Np, the rest entraining with the UF_6.

29-4

There is a slight difference in volatility between the NpF_6 and UF_6, which is enough to result in more NpF_6 than UF_6 remaining behind when the contents of a bottle are fed into the cascades. Thus, the concentration of NpF_6 tends to build up in the "heel" as the bottle is used repeatedly. The fractional retention from a single filling is not known.

The NpF_6 passes into the cascades with the UF_6 but the differential in volatility at operating conditions (optimal for UF_6 enrichment) leads to fine deposits of NpF_6 in the tubes of the barriers and the inside surfaces of the cascade units. It is found more often at the distal end of a system of cascades and in the U^{235} channels. This can't be predicted with certainty since some units will have much NpF_6 and some none. Probably about 60% of the NpF_6 deposits out here.

The problem arises when one of these cascade units is taken out of its operation sequence and opened for replacement of barriers. There is a definite program for such restoration and in some cases they are replacing the old barriers with new ones of improved design. These cascade units are housed in thick stainless steel tanks about 12 feet in diameter and 15 feet tall; they are welded shut and in general much too large to be handled by conventional industrial hygiene measures. The units have to be moved with an overhead travelling crane, special multi-wheel trucks, etc.

The units must be cut open with torches to get at the barrier tubes; the pieces certainly can't be handled gently or contained very readily because they are too massive.

The workers are supposed to wear special MSA nose-mouth face masks but they are not controlled too closely--I watched one man push up his mask and smoke a cigarette using potentially contaminated hands and gloves. They have devised some air-scoopes to fit around the ends of the unit as it is being torched open, but I would judge them to be of limited effectiveness. There may be a filter on the exhaust line for this air collector but it was not obvious; the exhaust simply dumps air outside the building.

Nevertheless, this ventilation was said to be very helpful. Fortunately, NpF_6 does not diffuse very readily, it having been found only within 8 feet of where the cascade unit had been cut free or opened.

According to Handbook 69 calculations (where the 200-year biological half-life is used), the MPC is 8.8d/min/m^3 of air. There are 1526 d/min/μg of Np^{237} so that the MPC equals about 0.0066 $\mu g/m^3$ of air.

Also, using the figures in Handbook 69, the maximal body burden would come to 1.3 d/m/24 hr urine sample coming from an 87.3 μg deposit. This, however, is so impossible that they have been using 13 d/m/24 hr urine sample as their standard on the basis that the true body content, after being out of contact with Np for 6 months to a year, would be 10% of the 48 hour lay-off concentration. (I think I am reporting their logic correctly.) Furthermore, the solubility of Np^{++++} is quite different from Np^{++++++} and it is not known which solubility factor should be used in the calculations.

* *

Np^{237} has a $T_{\frac{1}{2}}$ of 2.2 x 10^6 years and omits α-particles [illegible] 4.77 mev which can't be differentiated from other probable α-emitters by pulse height analysis. Gamma rays are also emitted and the 27.4 day daughter Pa^{233} emits both β- and γ-rays which are [illegible].

Recovery of Np from biological samples is poor (80% or [illegible] variable) and Dr. Levin reported trouble in making up standard solutions at concentrations of 10^{-11} or 10^{-12} where the biological [illegible] are supposed to fall, a situation similar to the NBS Ra-Th standard distributed by Dr. Beard.

Np^{239} with its 2.3 day $T_{\frac{1}{2}}$ and γ-rays is useful for some work but would not be satisfactory for chronic biological experiments.

Np^{237} can now be detected in urine but not consistently and it is [illegible] not very reliably. With their present techniques, the average is 0.22 d/m/24 hr urine sample for 75 people. The highest was 11 d/m/24 hr sample. Their spiked blank samples ran 25% to 75% of the expected values.

There are possibly 300 people at Paducah who should be checked out but they hesitate to precede to intensive studies because of the union's use of this as an excuse for hazard pay.

The whole body burden for Np^{237} by Handbook 69 is 6 x 10^{-8} curies and tests with a Np^{237} source on the Y-12 whole body counter put 7 x 10^{-9} curies as the counting limit so that the whole body counter may have some usefulness. They (Dr. Ward and others) were not receptive to the idea of sending 8 or 10 of the men with highest urine counts to Y-12 for counting.

I pointed out that we were planning to initiate biological distribution and radiotoxicological studies of Np^{237} which might have the effect of changing the MPC and burdens, but it would be two years or more before the data would be available. In view of that, I

urged both Dr. Ward and Mr. Stiller to improve the industrial hygiene measures surrounding the reworking of the cascade units. I don't have too much faith in masks and the dust particles here are about 0.5μ, the very worst size, biologically speaking.

I also pointed out to Dr. Ward the need to get post mortem samples on any of these potentially contaminated men for correlation of tissue content with urine output, but I am afraid the policy at this plant is to be wary of the unions and any unfavorable public relations.

Dr. Levin seems to be one of the authorities in the field of Np chemistry (others are: Weinstock, ANL; Eugene Lamb, ORNL, George Boyd, ORNL) and is interested enough to want to continue with efforts to improve the bioassay techniques. When he succeeds in this we may be better able to tackle this problem of whole body counting.

The potential situation at Paducah is enough to warrant radio-toxicological studies. In addition, there are the requirements for Np^{237} for various devices and the exposures during separation procedures at the Hanford OPP; perhaps Savannah River also is separating this isotope. I was told that the chemical separation of Np from U is very satisfactory, but the human factor in handling gram amounts should be considered a source of potential exposure.

Thus, it appears that Paducah has a Np problem but we don't have the data to tell them how serious it is. They may get into difficulties with the present Handbook 69 numbers and the problem of the body burden will inevitably come more to the forefront.

cc:
Dr. C. S. Shoup, ORO
B. Stiller, Paducah, thru C. S. Shoup, ORO
Director, Division of Production
Director, Office of Health & Safety

HDBruner:IMP

3-11-60